Ceramic Composite Hot Gas Filter Development

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1.0 INTRODUCTION

The focus of this paper is on the fabrication and testing of full size ceramic composite filter elements. The results of sub-scale testing performed in 1995 were used to identify a starting filter composition for full-scale filter fabrication and testing portion of the program. The work included the scale up of the filament winding process to produce 1.5 meter filter elements; filter improvement/cost reduction; and finally, initiation of the production of 50 filters for testing.

2.0 BACKGROUND INFORMATION

A critical feature of advanced coal fired power generation systems such as pressurized fluid bed combustors (PFBC) and integrated gasification combined cycle (IGCC) is the high temperature high pressure gas stream utilized by the gas turbine. In order to protect the gas turbine components from erosion, it is necessary to remove the ash/sorbent particulates from the turbine inlet gas stream. In first generation PFBC plants such as Tidd, hot cyclones provided a sufficiently clean gas stream for the ruggedized turbine. Second generation combined cycle plants utilize a topping combustor and high temperature gas turbines that require barrier filters to meet the turbine inlet requirements. The high temperature barrier filters are therefore considered to be one of the enabling technologies for the high efficiency cycles. Testing at various DOE and private facilities has demonstrated that the level of mechanical durability exhibited by the currently available filters may not be adequate to meet the reliability demands of large power generation systems.

3.0 PROGRAM OBJECTIVES

The objectives of this program are to develop toughened ceramic hot gas filters and evaluate these filters for application in PFBC and IGCC power generation systems.

4.0 APPROACH

The essential requirements of a composite material designed to meet the program objective for a toughened hot gas filter include the following:

• stable continuous fiber
•engineered fiber coating (if required)
• rigid porous matrix
• cost effectiveness

Structural reinforcement is provided by the continuous ceramic fibers while the discontinuous fibers perform the filtration function and form the rigid porous matrix of the filter element. A modified filament winding process was developed that simultaneously deposited both continuous and discontinuous fibers on a porous mandrel. An En-Tec computer controlled filament winder was used for all samples. As shown in Figure 1, the filter element preform is produced by winding the continuous fiber, Nextel 610, onto a porous vacuum mandrel. As the continuous fiber is being wound, a dilute slurry of Saffil fiber is pumped onto the mandrel. The excess water from the slurry was removed by the vacuum system to deposit the Saffil. The pure alumina Nextel 610 fiber was supplied as 400 filament tows with a unit weight of 1500 denier. The Saffil discontinuous fiber (95-97% alumina, 3-5% silica) was supplied as bulk fiber and exhibited a mean diameter of 3.5 microns. The resulting preform exhibits a continuous distribution of Nextel continuous fiber and Saffil discontinuous fiber through the wall thickness. The relative distribution of the continuous and discontinuous fibers was controlled by varying either the winding speed or the slurry feed pump speed. In addition, the relative distribution of the two fibers may be varied during the winding to optimize the cost and/or performance of the filter element.

The fabrication process is completed by the addition of a bond component in the form of a sol or liquid chemical binder to the filter element preform followed by heat treatments to convert the sol/chemical binder to a stable bond phase. An ideal bond system must develop bonds at fiber to fiber contact points without plugging or filling the open or continuous porosity of the filter element. The development of a uniform distribution of the bond phase is critical to developing high strength without compromising the permeability of the filter.

Phosphoric acid has been utilized as a bond system in the refractory industry for many years and details of the bonding reactions are given in the literature[1]. The use of phosphoric acid as a binder for metal matrix composite fiber preforms has also been reported[2]. Phosphoric acid reacts with the alumina fiber to form an aluminum metaphosphate gel on the fiber surface which converts to the final bonding phase, $AlPO_4$, on heating. Bonding occurred at the fiber to fiber contact points with minimal filling of the porosity of the fiber preform. Because phosphoric acid reacts with any source of alumina, the continuous fiber in the preform must be coated with an inert material such as carbon to minimize the reaction and the associated strength degradation. In addition, all heat treatments of the phosphoric acid bonded filter elements must be conducted in an inert atmosphere to protect the carbon coating because the continuous fibers are vulnerable to the phosphate decomposition products. The use of uncoated continuous fibers with the phosphoric acid bond system resulted in brittle failures with little or no advantage over monolithic filter element systems.

An oxyhydroxide of aluminum, AlOOH or boehmite was included as an alternative to the phosphoric acid bond system. On heating, the boehmite transformations follow the sequence:

$$AlOOH \rightarrow \gamma\text{-}Al_2O_3 \rightarrow \delta\text{-}Al_2O_3 \rightarrow \theta\text{-}Al_2O_3 \rightarrow \alpha\text{-}Al_2O_3$$

The initial transformation from the oxyhydroxide to alumina occurs between 450 and 580 °C depending on the crystallinity of the starting material. Filter elements typically exhibited non-uniform binder distribution with the highest concentration occurring near the outer surface of the element. This type of binder distribution was attributed to migration of the liquid sol to the hot outer surface of the element where the excess water evaporated causing an accumulation of the boehmite bond phase. A more uniform distribution of the bond phase should allow increased binder contents and the associated higher strength with less of an impact on the permeability. No fiber coating is required because the boehmite sol does not react with the Nextel fiber.

Two separate filter element evaluations were performed in the Westinghouse high temperature, high pressure test facility in Pittsburgh, PA. Each test included simulated PFBC steady state pulse cycling conditions, accelerated pulse cycling, and thermal transient conditions. Phosphoric acid bonded and boehmite bonded Nextel 610/Saffil composite filter elements were tested in August, 1996 and April 1997, respectively.

Sample filter elements were characterized in terms of their microstructure, permeability, and mechanical properties. The permeability of test specimens was determined from the face velocity and the associated pressure drop across the specimen. Compressive C-ring tests were performed on a computer controlled mechanical test machine using calibrated load and deflection sensors. All C-ring testing was performed at 1600 °F. Five one inch wide C-rings were tested from each filter element.

The distribution and relative amounts of continuous fiber, chopped fiber, and bond phase was determined by examination of polished sections in an Etech scanning electron microscope (SEM) equipped with energy dispersive spectroscopy (EDS). In order to minimize damage during sample preparation, samples were vacuum impregnated with a thermal plastic and polished. The thermal plastic was dissolved in a solvent and then heated in air to remove the residual resin. The resulting sample provided an unaltered structure for SEM examination.

5.0 RESULTS AND DISCUSSION

Filter element compositions and fiber architectures are given in Table 1 for the different tests. Specific composition and permeability data of the filter elements from the W-STC test of 8/96 are summarized in Figure 2. These elements utilized phosphoric acid as the bond system and carbon coated Nextel 610 fiber. The pressure drop at a face velocity of 10 ft/min exceeded the Westinghouse specification of 10 in-wg. Typical high temperature C-ring results for these filter elements in the "as fabricated" condition and post-test condition are given in Figure 3. The filter elements exhibited a retained strength of approximately 70% with non-brittle fracture behavior. The distribution of Nextel 610 and Saffil on the outside surface of element 6-6-6 is shown in Figure 4. The cross-section of the filter element is shown in Figure 5. Sealing problems encountered in two of the four test elements was attributed to the flange machining operation which cut the continuous fibers in the region of the highest collaring loads.

Based on these results, a filter improvement and cost reduction task was added to the program. This task was directed at optimization of the Nextel 610 fiber shape and architecture; further development of the boehmite bond system; and development of a net shape flange.

As indicated above, the ability to control the relative amounts of continuous and discontinuous fiber makes it possible to optimize the distribution of the highest cost raw material, the Nextel 610 continuous fiber. Figure 6 shows the ratio of Nextel 610 to Saffil as a function of the number of layers or closures from the inside diameter to the outside diameter of the filter element. This demonstrates how the Nextel 610 was concentrated in regions of high stress near the filter element ID and OD and decreased near the center of the filter element wall. An improved method of adding the boehmite bond sol was also incorporated in this filter. The resulting C-ring strength of this filter element is shown in Figure 7.

A second filtration test in the W-STC high temperature, high pressure test facility was performed on the improved filter elements in 5/97. Figure 8 shows the relative amounts of Nextel 610, Saffil, and binder for the filter elements submitted for this test. Also included on Figure 8, is the pressure drop data which demonstrates the effectiveness of the improved fiber architecture. The results of characterization tests for these samples will be reported at a later date.

A summary of the composition and permeability data for the Karhula samples is shown in Figure 9. These data also provide an indication of the consistency of the fabrication process.

The overall status of the project is summarized in Table 2 which compares the filter requirements to the current status of the McDermott Techology Inc. filter manufacturing process. The primary challenge will be to scale up the manufacturing process to realistic production levels at acceptable costs.

6.0 BENEFITS

This program has demonstrated a hot gas filter concept and a flexible fabrication method that resulted in an oxide-oxide composite based filter with improved strength and toughness compared to monolithic filter materials. In addition, the flexibility of the process in terms of the fiber distribution should result in improved performance and cost.

7.0 FUTURE ACTIVITIES

The production of 50 filter elements for testing in DOE demonstration facilities is in progress. The performance of these elements in the Karhula test will be used to guide future filter element improvements. The fabrication process described above will be continuously evaluated to identify scale-up issues and areas where automation will improve product quality and cost. The upcoming Lakeland project will require a significant investment in production equipment to meet the schedule.

8.0 ACKNOWLEDGMENTS

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9.0 REFERENCE

1. W. D. Kingery, “Fundamental Study of Phosphate Bonding in Refractories: I, Literature Review”, J. Am. Cer. Soc., 33, (8) 239-241, (1950).

2. Chiou, Jeng-Maw and D. D. L. Chung, “Improvement of the temperature resistance of aluminum-matrix composites using an acid phosphate binder”, J. Mat. Sci. 28, 1435-1446, (1993).

Table 1. Filter element composition and fiber architecture.

	W-STC 8/96	W-STC 5/97	Karhula 8/97
continuous fiber	Nextel 610	Nextel 610	Nextel 610
chopped fiber	Saffil	Saffil	Saffil
continuous to chopped ratio	2:1	variable	variable
continuous fiber coating	carbon	none	none
continuous fiber architecture	45º helical	45º helical	45º helical
bond type	phosphoric acid	boehmite	boehmite
flange	machined	net shape	net shape

Table 2. Hot Gas Filter Summary

Property	Req't.	Status	Challenge
size	2.4 x 60"	2.4 x 60"	complete
shape	flanged, closed end tube	flanged, closed end tube	complete
pressure drop @10ft/min	10	5	complete
strength	1 - 4 ksi	0.8 - 1.7 ksi	moderate
toughness	non-brittle failure	non-brittle failure	moderate
thermal shock	ambient air back pulse	70% retained strength	moderate
corrosion resistance	3 year life	tbd	significant

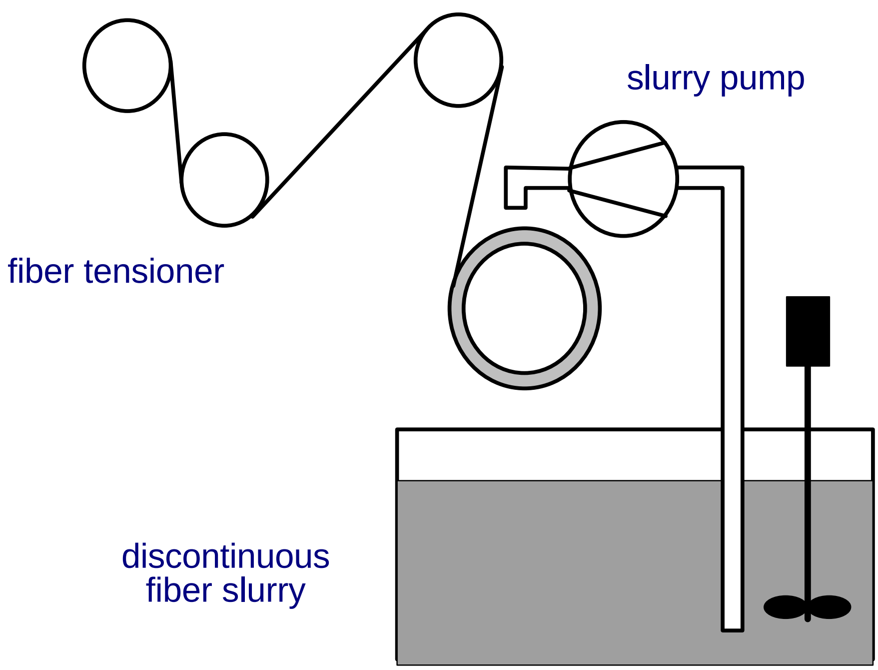

Figure 1. Schematic diagram of vacuum winding process.

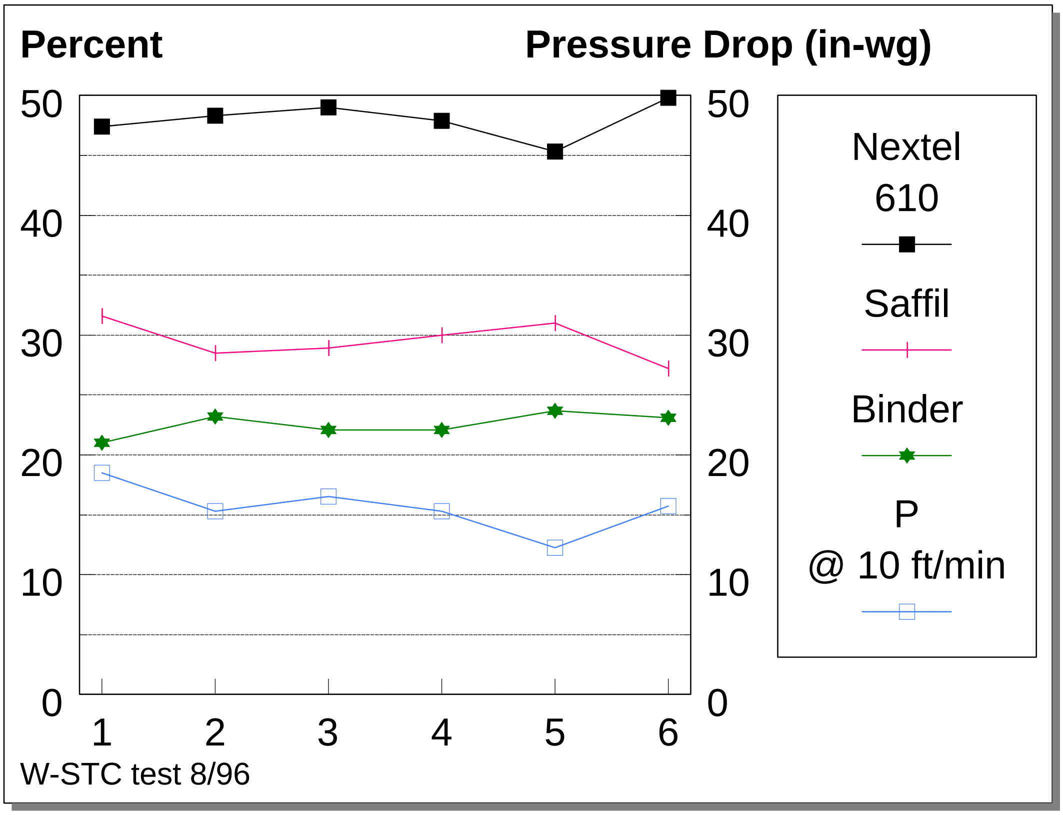

Figure 2. Filter element composition and permeability for W-STC test 8/96. (Preliminary data)

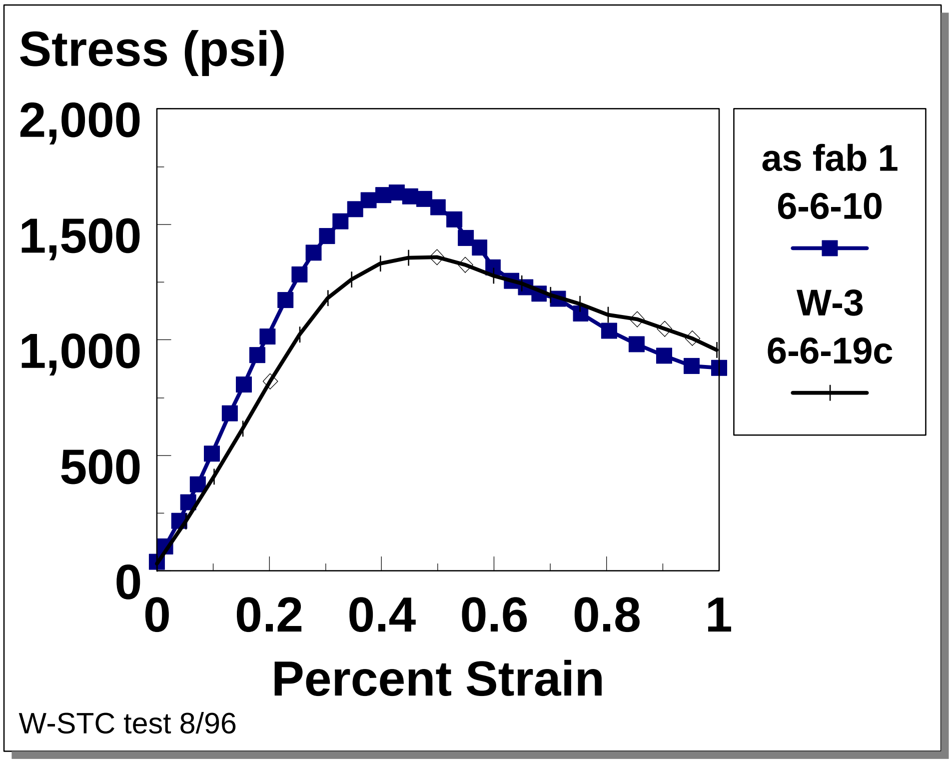

Figure 3. As-fabricated and post-test C-ring results for W-STC 8/96 test. (Preliminary data)

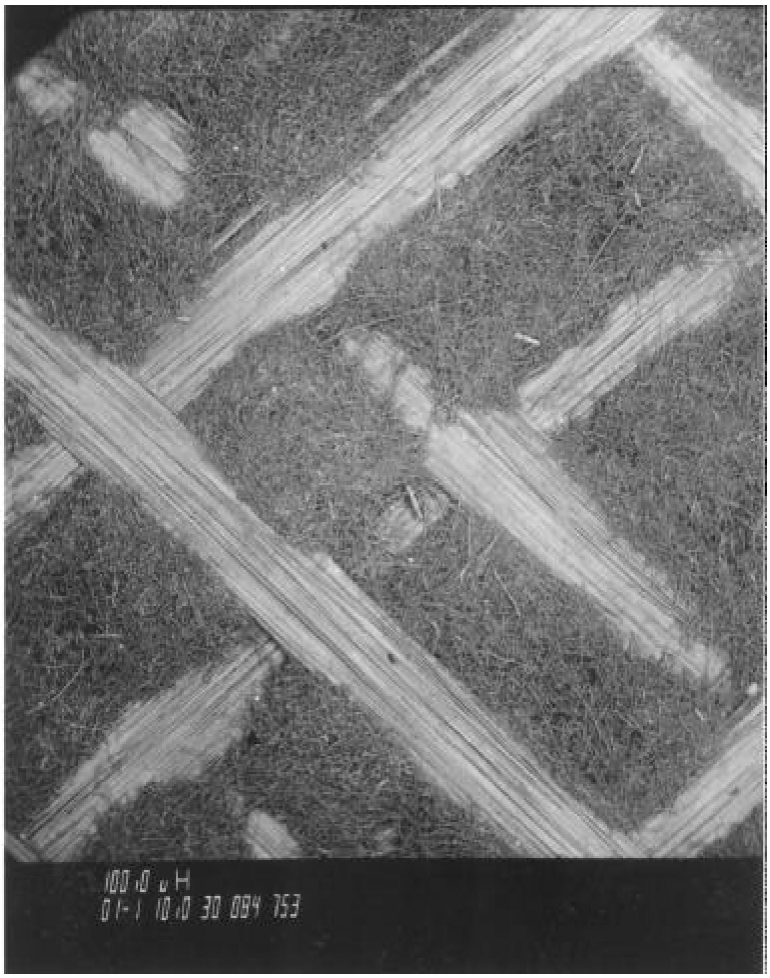

Figure 4. Outside surface of sample 6-6-6 following 8/96 W-STC test.

Figure 5. Cross-section of sample 6-6-6 showing distribution of Nextel 610 and Saffil fibers.

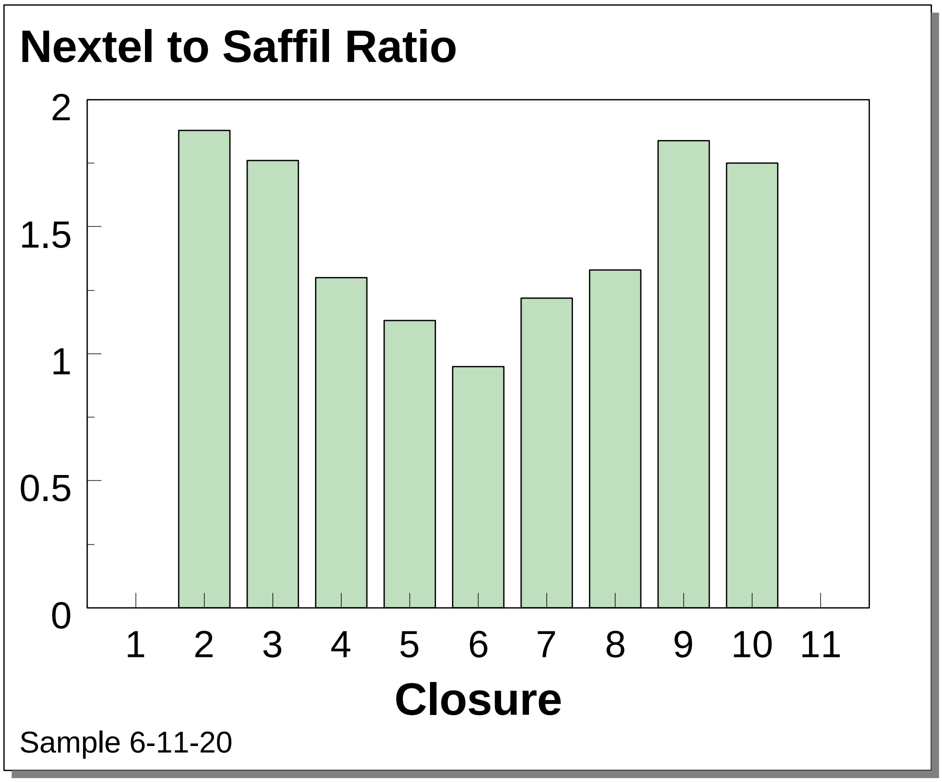

Figure 6. Example of controlled distribution of Nextel 610 fiber. (Preliminary data)

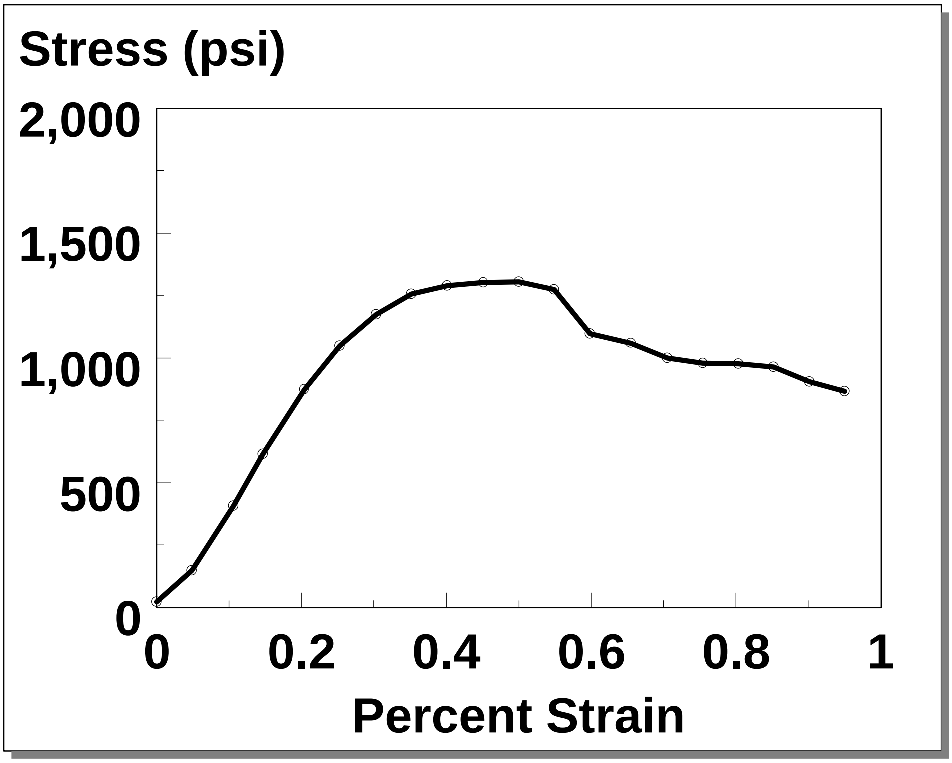

Figure 7. C-ring strength of sample 6-11-20 with controlled fiber distribution. (Preliminary data)

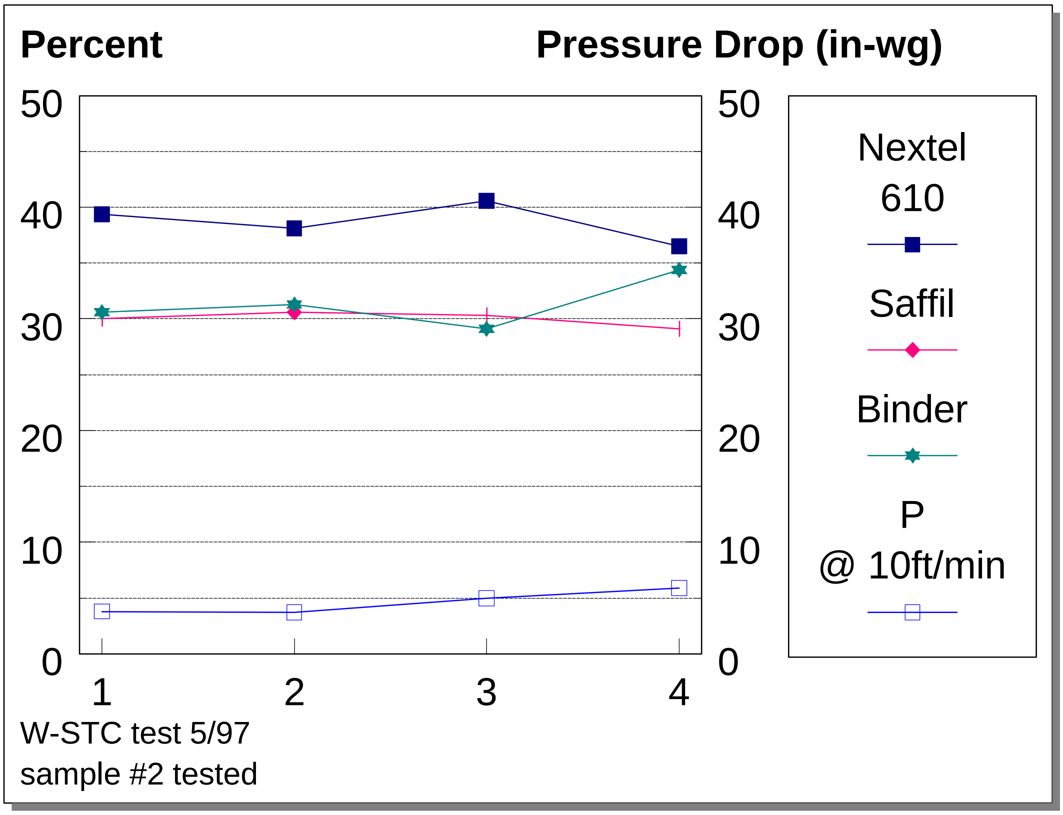

Figure 8. Filter element composition and permeability for W-STC test 5/97. (Preliminary data)

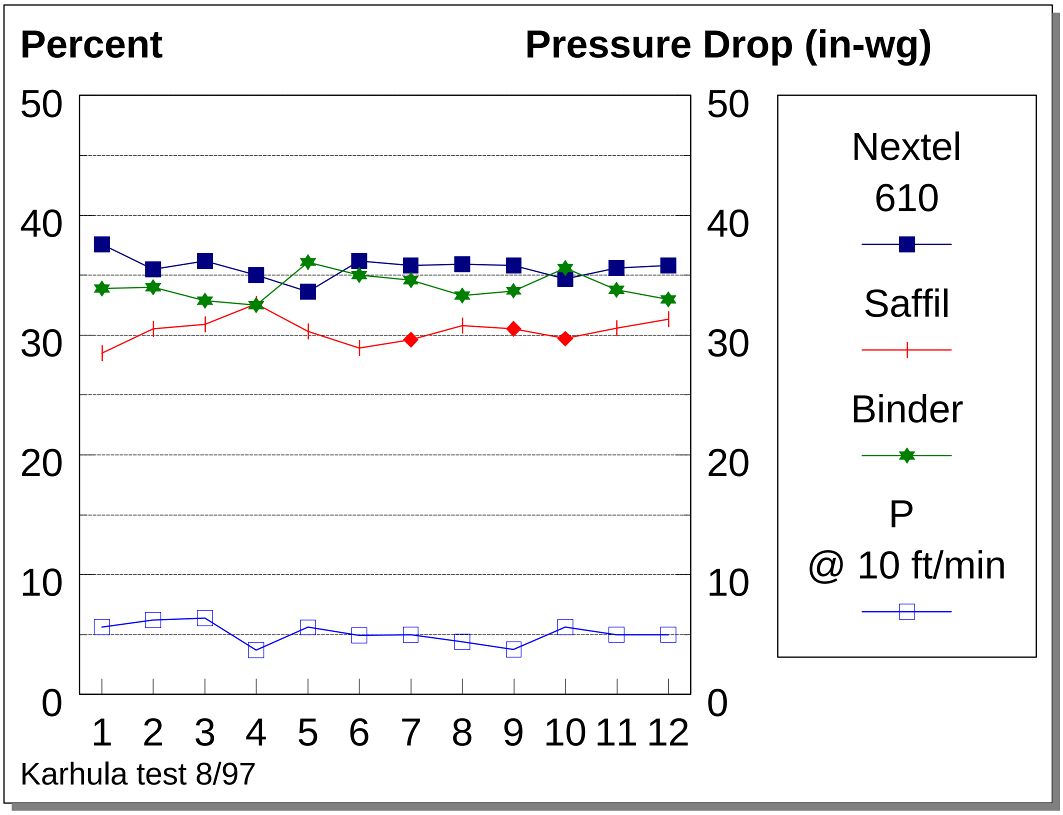

Figure 9. Filter element composition and permeability for Karhula test elements. (Preliminary data)